Nuclear Magnetic Resonance in a Ferromagnet-Semiconductor Heterostructure

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We report the observation of nuclear magnetic resonance (NMR) in a ferromagnet-semiconductor heterostructure in the presence of a spin-polarized current. Spin-polarized electrons injected from a metallic ferromagnet generate a large nuclear spin population in a GaAs quantum well by dynamic polarization. The characteristic time for the polarization process is approximately 20 sec, and the nuclear polarization can persist for several minutes after the current is turned off. Resonant depolarization is observed in the presence of an AC magnetic field or when the injection current is modulated at the NMR frequency.

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A detailed understanding of the hyperfine interaction is a prerequisite for the coherent control of electron and nuclear spin systems in semiconductor heterostructures. Electrical spin injection[1] provides a simple means of introducing spin-polarized carriers into a semiconductor, but it has proven more difficult to achieve the degree of tunability over the combined electron and nuclear spin systems demonstrated in optical pumping experiments.[2,3,4,5] For example, Poggio *et al.*[6] have recently shown that it is possible to electrically control the spatial profile of optically-pumped nuclear polarization within a single quantum well. One of the advantages of optical pumping is the efficiency of dynamic nuclear polarization (DNP), in which a non-equilibrium nuclear polarization is generated by the spin relaxation of electrons.[7,8,9,10] In principle, the DNP process can also be driven by a spin-polarized current.[11] This was inferred in recent experiments on $Fe/Al_xGa_{1-x}As$ heterosructures,[12] although an explicit dynamical signature of DNP was not demonstrated.

In this letter we report the direct observation of nuclear magnetic resonance (NMR) in a GaAs quantum well (QW) in the presence of a spin-polarized current. We demonstrate that DNP can be partially supressed by resonant depolarization of the nuclear spins. The depolarization can be achieved either by the traditional approach of applying a resonant AC magnetic field or by modulating the spin-polarized current at the NMR frequency. The time dependence of the DNP process is measured, indicating a characteristic build-up time of ≈ 20 sec and persistence of nuclear polarization up to eight minutes after the spin-polarized current has been turned off.

The heterostructure is grown by molecular beam epitaxy on a p^+ GaAs (100) substrate and consists of $p\text{-}Al_{0.1}Ga_{0.9}As/QW/n\text{-}Al_{0.1}Ga_{0.9}As/Fe/Al$. The QW comprises 100 Å GaAs between intrinsic $Al_{0.1}Ga_{0.9}As$ barriers, and the Fe film is 50 Å thick. The metal layers are grown *in situ* at $\approx 0°$ C, and a 3×10^{13} atoms/cm^2 δ-doped layer of Si is grown 25 Å from the $Fe/Al_xGa_{1-x}As$ interface to create a thin Schottky tunnel barrier. The devices are fabricated into bars with an active region 80 μm wide (see schematic drawing in Fig. 1), and the experiments are carried out in a magneto-optical cryostat at temperatures ranging from 2 K to 40 K.

The measurements are performed in the Voigt geometry with the applied field $\mathbf{H}_0$ in the plane of the QW. Electroluminescence (EL) is collected along the growth direction, indicated by $\mathbf{k}$ in Fig. 1, which coincides with the direction of the AC magnetic field $\mathbf{H}_1$ generated by a four-turn coil placed on top of the sample. The EL is due to the recombination of electrons tunneling from the Fe layer with heavy holes flowing from the substrate. The circular polarization of the EL, referred to as the ELP signal, is equal to the component of electron spin polarization along $\mathbf{k}$ at the time of recombination.

Typical ELP data as a function of applied field are shown in Fig. 1. All of the measurements under discussion here were conducted with $\mathbf{H}_0$ nearly parallel to the $[01\overline{1}]$ direction, which corresponds to a magnetic hard axis. As H_0 decreases below 500 Oe, the magnetization $\mathbf{M}$ rotates away from $[01\overline{1}]$ towards the [011] (easy) direction. The non-zero angle between the injected spin $\mathbf{S}_0 \parallel \mathbf{M}$ and $\mathbf{H}_0$ leads to precession of the spin in the QW, resulting in a steady-state component of $\mathbf{S}$ along $\mathbf{k}$ and hence the observed ELP signal. The ELP hysteresis loops shown in Fig. 1 can be modeled extremely well provided that the total field $\mathbf{B} = \mathbf{H}_0 + \mathbf{B}_N$, where $\mathbf{B}_N$ is assumed to be the hyperfine field due to nuclei polarized by DNP. As discussed in Ref. 12, the magnitude of $\mathbf{B}_N$ depends on the injection current density and the angle θ between $\mathbf{S}_0$ and $\mathbf{H}_0$ and reaches several kilogauss for current densities ~ 10 A/cm^2 and $\theta < \pi/2$.

The large nuclear magnetic field is due to the long spin-relaxation time for nuclei and the large difference between the electron and nuclear magnetic moments.[10] Since the nuclear spin-lattice relaxation time can be many minutes,[2,13] we expect the field $\mathbf{B}_N$ to build up on laboratory time scales when the current is turned on and then decay slowly when the current is turned off. Figure 2 shows the ELP signal as the device current is switched on and off for variable amounts of time. In this case, the current is off for ten minutes prior to the start of data collection at $t = 0$. The LED is then turned on

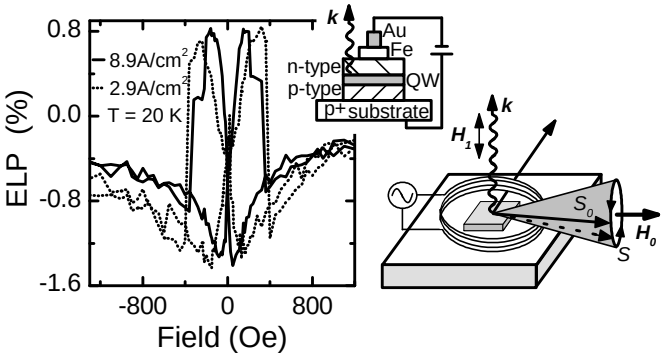

FIG. 1: Electroluminescence polarization as a function of the applied field H_0 for two current densities. Higher current corresponds to larger nuclear field. Center inset: cross-sectional schematic of the device. Right inset: schematic of the experimental setup. The device is at the center of a coil which generates an AC magnetic field $\mathbf{H}_1$ out of the plane, parallel to the direction of light propagation $\mathbf{k}$. $\mathbf{H}_0$ is in the device plane. The electron spin $\mathbf{S}$ precesses about $\mathbf{H}_0$ after injection from Fe into the GaAs. The angle between the initial electron spin $\mathbf{S}_0$ and $\mathbf{H}_0$ is determined by the anisotropy of the Fe film.

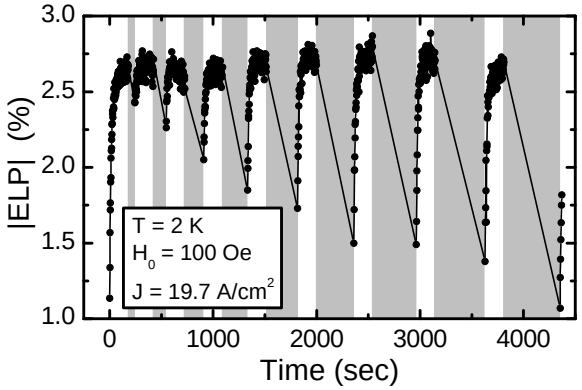

FIG. 2: Laboratory time dependence of the electroluminescence polarization (ELP) signal. The device current is left off for ten minutes prior to $t = 0$, and the current is then switched between on (three minutes) and off (gray regions), with the $t_{\rm off}$ increasing from 1 minute to 9 minutes.

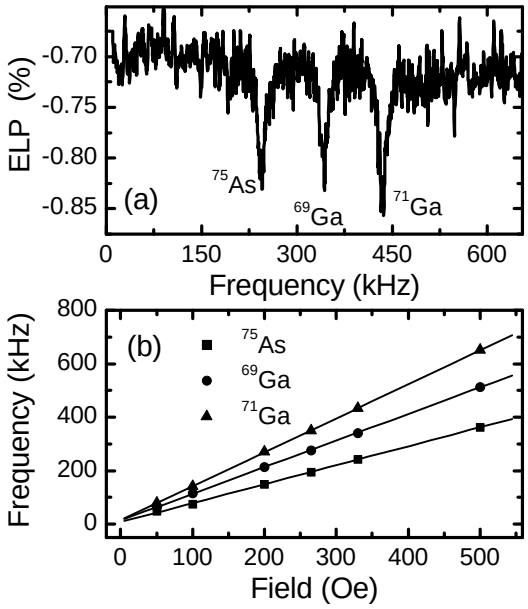

FIG. 3: (a) Frequency-swept NMR depolarization features at T = 20 K, $H_0 = 330$ Oe, and $H_1 \approx 0.2$ Oe, corresponding to the three isotopes present in the GaAs QW. (b) Depolarization peak frequency as a function of H_0 at 20 K and $H_1 \approx 0.2$ Oe. The solid lines are linear fits.

and the ELP signal is measured for three minutes. The increase in the ELP signal is due to the increase in total internal magnetic field and the consequent increase in the angle of precession of the injected spin polarized electrons. To probe the lifetime of the nuclear polarization in the absence of continuous pumping, the current is turned off for a time $t_{\rm off}$ and then on again for three minutes. Figure 2 shows a series of these measurements with $t_{\rm off}$ varying from 1 min to 9 min. The first data point taken after each "off" period indicates the degree of nuclear polarization that has been retained. By repeating this procedure with increasing $t_{\rm off}$, we observe a decay time of approximately 5 min. Although both the build-up and decay times for the ELP signal are long, it is difficult to make a direct quantitative mapping between the magnitude of the ELP signal and the actual nuclear polarization. In particular, the relationship between the ELP signal and B_N is very non-linear. The ELP signal increases rapidly at small nuclear fields and then saturates,[12] and this is responsible in part for the large difference between the build-up and decay times for the ELP signal.

Although the nuclear polarization can relax slowly through coupling to the lattice, the most effective way of destroying DNP is to apply an oscillating magnetic field $\mathbf{H}_1$ at the NMR frequency along a direction perpendicular to $\mathbf{H}_0$. The transverse field couples directly to the nuclear spin system, leading to depolarization of the nuclei that are in resonance. H_1 is generated by a 1 cm diameter, four turn coil driven by the sinusoidal output of a function generator. By sweeping the frequency of H_1 at fixed H_0, depolarization signatures for each of the three isotopes in the GaAs QW [^{75}As ($\gamma = 0.731$ kHz/Oe), ^{69}Ga ($\gamma = 1.025$ kHz/Oe), and ^{71}Ga ($\gamma = 1.302$ kHz/Oe)][14] are observed, as seen in Fig. 3(a) for $H_0 = 330$ Oe. In each case, the linewidth of the depolarization signature is 10 - 15 kHz, with no systematic dependence on magnetic field. The linewidth at these fields (< 500 Oe) is therefore not limited by inhomogeneous dephasing.

Frequency sweeps were conducted at several different magnetic fields. The peaks of the depolarization features for all three isotopes are plotted versus H_0 in Fig. 3(b). Linear fits of $\nu = \gamma H_0$ were obtained for each isotope, yielding gyromagnetic ratios $\gamma_{^{75}\rm As} = 0.71 \pm 0.1$ kHz/Oe,

$\gamma_{^{69}\mathrm{Ga}} = 1.00 \pm 0.04$ kHz/Oe, and $\gamma_{^{71}\mathrm{Ga}} = 1.27 \pm 0.04$ kHz/Oe, in agreement with the accepted values. The deviations from the fits are smaller than the limit set by trapped flux in the magnet (≈ 10 Oe).

The sign of the depolarization features warrants comment, since the dips in Fig. 3(a) correspond to an increase in the magnitude of the ELP signal, which is negative in this field range. We have confirmed that the features in Fig. 3(a) correspond to a *decrease* in the nuclear polarization by comparing with the observed dependence of the ELP signal on current density.[12] The reason for this non-intuitive result is that for sufficiently large nuclear fields, the average angle through which the electrons precess during their lifetime becomes larger than $\pi/2$. An increasing nuclear field therefore results in a decrease in the magnitude of the ELP signal because the electrons are actually precessing further away from the measurement axis. Although a complete quenching of B_N would result in nearly total suppression of the ELP signal, only a single isotope is depolarized at each resonance. We have confirmed that for low B_N, corresponding to applied fields less than 60 Oe, resonant depolarization results in a decrease in the ELP magnitude as expected.

Since the nuclear polarization in these devices is generated by the injected electrons, an alternative approach to depolarization is resonant modulation of the device current. In this case, the coil is not used ($H_1 = 0$), and the DC bias voltage is set above the threshold for light emission. An AC voltage is then added so that the average bias voltage corresponds approximately to the value used in the measurements discussed above. In this case, the bias is being used only to modulate the injection current. Given the square well design, we cannot control the relative position of the nuclear and electronic spins as achieved recently in parabolic quantum wells.[6] Fig. 4 shows the ELP signal as a function of bias voltage frequency for $H_0 = 330$ Oe, $V_{DC} = 2.1$ V, and $V_{AC} = 0.5\ V_{PP}$. The principal resonances are clearly visible, as can be seen by comparing the data taken under alternating device current (lower curve) with the data taken under AC magnetic field (upper curve). In the case of alternating device current, however, there are many other peaks in the data, and frequency sweeps at different fields indicate that these also correspond to nuclear resonances. Some of the additional peaks can be mapped to sub-harmonics of the primary resonances, such as $\nu = \gamma H_0/3$. Other peaks occur at either harmonics of the primary resonances, such as $\nu = 2\gamma H_0$, or their sub-harmonics, such as $\nu = 2\gamma H_0/3$. Similar harmonic and subharmonic resonances have also been observed in quantum well systems with quadrupolar interactions in which a strong electric field gradient is modulated with a gate voltage.[6]

Sub-harmonics of the primary resonances are never observed with an AC magnetic field, and we attribute them to the non-sinusoidal time dependence of the injection current as the bias voltage is modulated. This is a consequence of the diode structure of the device. The origins of harmonics of the fundamental resonances are less clear. These peaks are also observed in AC magnetic field experiments at high amplitudes ($H_1 > 0.4$ Oe). The observation of the $\Delta m = \pm 2$ peaks in the AC magnetic field experiments at high amplitudes indicates that in this experiment they originate from some intrinsic non-linearity. Dipole-dipole coupling can lead to both $\Delta m = \pm 2$ and $\Delta m = \pm 3$ transitions,[15] and may play a more significant role because of the large nuclear polarization in these experiments.

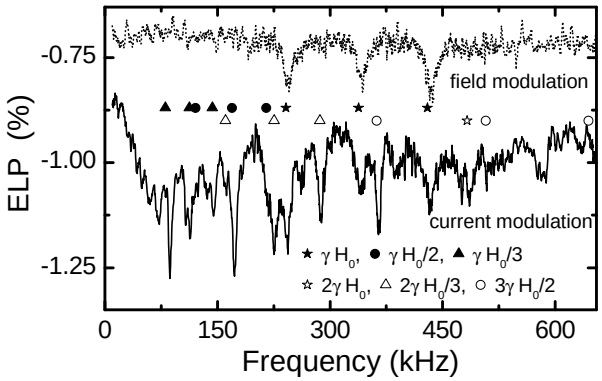

FIG. 4: Frequency-swept NMR depolarization features at T = 20 K, and $H_0 = 330$ Oe corresponding to the three isotopes present in the GaAs QW. The upper curve shows the ELP signal measured with an AC magnetic field $H_1 \approx 0.2$ Oe. The lower curve shows the ELP signal measured under alternating device current ($H_1 = 0$ Oe). The filled symbols identify principal nuclear transition frequencies and their subharmonics. The empty symbols indicate harmonics of the primary resonances as well as their sub-harmonics.

The spectroscopic measurements presented here unambiguously demonstrate dynamic nuclear polarization by electrical spin injection. The ability to modulate the nuclear polarization with a spin-polarized current provides a unique means of controlling the coupled electron-nuclear spin system. This capability, in combination with the large nuclear polarization achieved at low fields, provides a new approach to NMR spectroscopy of heterostructures.

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